Crossed Andreev Reflection in Diffusive Contacts

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(Dated: February 6, 2008)

Crossed Andreev reflection in multiterminal structures in the diffusive regime is addressed within the quasiclassical Keldysh-Usadel formalism. The elastic cotunneling and crossed Andreev reflection of quasiparticles give nonlocal currents and voltages (depending on the actual biasing of the devices) by virtue of the induced proximity effect in the normal metal electrodes. The magnitude of the nonlocal processes is found to scale with the square of the barrier transparency and to decay exponentially with interface spacing. Nonlocal cotunneling and crossed Andreev conductances are found to contribute equally to the nonlocal current, which is of relevance to the use of normal metal-superconducting heterostructures as sources of entanglement.

PACS numbers: 03.67.Mn, 73.23.-b, 74.45.+c, 74.78.Na

I. INTRODUCTION

In standard Andreev reflection at a single normal metal-superconductor interface an electronlike quasiparticle in the normal metal can be transformed into a hole-like quasiparticle of opposite momentum.[1] When two normal metal (N) electrodes or ferromagnets (F) are attached to a superconductor (S) at a distance from each other of the order of the coherence length, two additional nonlocal processes are possible. During elastic cotunneling (EC), an electron is transferred from one electrode to the other, while for crossed Andreev reflection (CAR) an electron in one of the electrodes is transformed into a hole in the other electrode.[2,3]

Bell-inequality experiments, quantum computation, and teleportation of quantum states require quantum objects that are entangled. Cooper pairs in superconductors are spin singlets and are, therefore, suitable sources of entanglement. Crossed Andreev reflection is a promising possibility for the creation of locally separated entangled electrons.[4,5,6,7,8]

Nonlocal transport properties have been seen experimentally in NS[9,10] and FS heterostructures.[11,12] The microscopic origin of the effects of EC and CAR, as well as possible ways in which these can be used for the production of locally separated entangled quasiparticles, remain to be understood theoretically.

Recently, theoretical studies of CAR have been performed for various types of junctions.[13,14,15,16,17,18,19] Modeling of nonlocal effects by means of perturbation theory using an effective tunnel Hamiltonian[13,17] has indeed provided EC and CAR, the signal being, however, vanishingly small (of the order of the fourth power in interface transparency). Consequent pioneering efforts to include disorder[14,19] and weak localization[16] have found enhanced effects, but still not to the level of experimental observations, so that the question as to the microscopic origin of CAR still remains open.

In this paper, we provide insight into the underlying microscopic mechanism of CAR by studying nonlocal transport by means of quasiclassical kinetic theory.

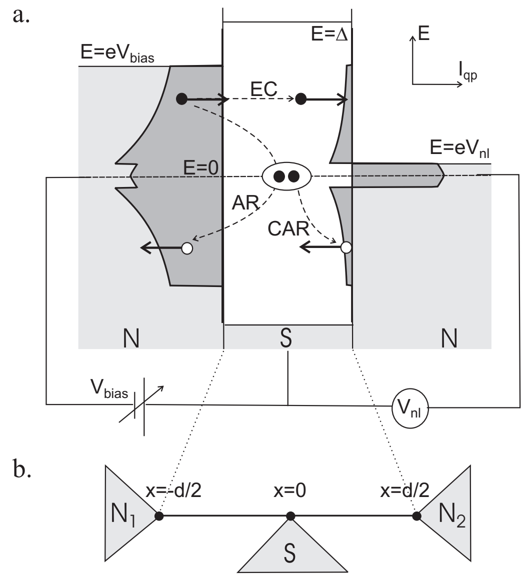

FIG. 1: (a) Schematic representation of the double-barrier NSN structure, which is voltage biased accross the first interface. An incoming electron from the left normal metal electrode, can undergo three different processes that contribute to the current: Andreev reflection (AR), elastic cotunneling (EC) and crossed Andreev reflection (CAR). The depicted spectral currents that result from these processes are the main result of this paper. (b) Results have been obtained from a one-dimensional Keldysh-Usadel quasiclassical Green's function calculation of the depicted structure.

We present a mechanism in which CAR exists by virtue of the proximity induced superconducting correlations in the electrodes, for arbitrary barrier transparency. The proximity effect is the essential new ingredient in our model, giving a large contribution to CAR, of second order in transparency, in contrast to the nonlocal effects of tunnel Hamiltonian models. We show how CAR relates to the competing process of co-tunneling. For ballistic transport, Andreev reflection is understood most straightforwardly, but we have modelled the additionally

challenging case of diffusive transport, as experimentally often is the case. Our model is of relevance to the future design of experiments that are based on Andreev entanglers.

II. QUASICLASSICAL MODEL

The most generic model system to study nonlocal effects in superconducting structures is a three-terminal configuration consisting of a quasi-1D superconducting wire of length d, attached to normal reservoirs $N_{1,2}$ and a superconducting reservoir S which can be independently biased, see Fig. 1. This model is an extension to the earlier approach by Volkov *et al.*[20] that was used to calculate nonequilibrium transport properties of two-terminal $N'NS$ contacts. The electrodes $N_{1,2}$ are weakly coupled to the wire, while the reservoir S is in good electrical contact with the wire so that their electric potentials are equal. Morten *et al.*[19] addressed in their circuit model the role of the latter coupling strength between superconductor S and wire. They found nonvanishing nonlocal effects, but only in the case of weak coupling. In the case of zero resistance between wire and S (strong coupling), the circuit results coincide with the tunnel Hamiltonian results of a vanishing nonlocal signal. However, in the experiments often no barrier is present between wire and superconductor. Hence, here we study the regime of good electrical contact and equal potentials between S and the wire.

We assume that transport is diffusive (scattering length being smaller than other length scales) so that the quasiclassical kinetic theory in the dirty limit can be applied. The Keldysh-Usadel diffusion equation for the Green's function, in Keldysh-Nambu space, in the absence of time-dependencies, magnetic field and inelastic self-energy terms, can be written as

$$-\hbar\mathcal{D}\nabla\left(\breve{G}\nabla\breve{G}\right)=\left[iE\breve{\tau}_3+\breve{\Delta},\breve{G}\right], \qquad (1)$$

where $\breve{\tau}_3=\begin{pmatrix}\hat{\tau}_3 & 0\\ 0 & \hat{\tau}_3\end{pmatrix}$, $\hat{\tau}_3=\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}$,

$$\breve{G}=\begin{pmatrix}\hat{G}^R & \hat{G}^K\\ 0 & \hat{G}^A\end{pmatrix}, \breve{\Delta}=\begin{pmatrix}\hat{\Delta} & 0\\ 0 & \hat{\Delta}\end{pmatrix}, \hat{\Delta}=\begin{pmatrix}0 & \Delta\\ \Delta^* & 0\end{pmatrix},$$

E is the energy measured from the chemical potential, and $\mathcal{D}$ is the diffusion constant. The current is given by

$$I=\frac{1}{2eR_N}\int dE\,\mathrm{Tr}\left[\hat{\tau}_3\left(\hat{G}^R\nabla\hat{G}^K+\hat{G}^K\nabla\hat{G}^A\right)\right], \qquad (2)$$

where R_N is the normal state resistance.

The quasiclassical modeling of the diffusive transport through such a NSN structure can be splitted in solving the retarded and Keldysh parts of Eq. (1) respectively.

A. The proximity effect

In order to calculate the retarded part of the Green's function, $\hat{G}^R$, it is convenient to use the standard θ-parametrization, $\hat{G}^R(x)=\hat{\tau}_3\cos\theta(x)+\hat{\tau}_2\sin\theta(x)$. The function $\theta(x)$ is a measure of the superconducting correlations at a given point within the structure and θ satisfies the Usadel equation[21]

$$\mathcal{D}\frac{\partial^2}{\partial x^2}\theta(x)+2iE\sin\theta(x)=0. \qquad (3)$$

At the NS interfaces at $x=\pm d/2$, the function $\theta(x)$ satisfies the following boundary conditions[22]

$$\gamma_B\xi_N\frac{\partial\theta_N}{\partial x} = \pm\sin(\theta_S-\theta_N), \quad x=\pm d/2, \qquad (4)$$

$$\gamma\xi_N\frac{\partial\theta_N}{\partial x} = \xi_S\frac{\partial\theta_S}{\partial x}, \quad x=\pm d/2. \qquad (5)$$

where $\xi_{N,S}=\sqrt{D_{N,S}/2\pi T_c}$ are the coherence lengths and $D_{N,S}$ are the diffusion coefficients in N and S respectively. The proximity effect parameters γ and γ_B are defined as $\gamma_B=R_B/\rho_N\xi_N$ and $\gamma=\rho_S\xi_S/\rho_N\xi_N$, where R_B is the interface resistance and $\rho_{N,S}$ are the resistivities of the N and S metals. These parameters have a simple physical meaning[22]: γ is a measure of the strength of the proximity effect between the S and N metals, whereas γ_B describes the effect of the interface transparency. From here on, $\gamma_B\gg 1$ is assumed, corresponding to a small barrier transparency.

Solutions to the proximity effect problem in diffusive junctions have been extensively discussed in various regimes.[23,24] Generally, a minigap exists in N, of the order of the Thouless energy. In the considered case of bulk N ($d_N\gg\xi_N$), the minigap vanishes. The quasiparticle density of states is given by $\mathrm{Re}\,G=\mathrm{Re}(\cos\theta)$ and the pair density of states is defined as $\mathrm{Re}\,F=\mathrm{Re}(\sin\theta)$. It is straightforward to solve the Usadel equation (3) with the boundary conditions (4) and (5) numerically. The calculated densities of states $\mathrm{Re}\,G_N$ and $\mathrm{Re}\,F_N$ at the N side of an NS interface are shown in Fig. 2. It is seen that the quasiparticle spectrum in N is gapless while strong superconducting correlations exist at low E, described by $\mathrm{Re}\,F_N$. As will be shown below, the existence of a nonzero $\mathrm{Re}\,F_N(E)$ at the NS interface is an essential ingredient to our solution of the nonlocal conductance in a diffusive NSN structure.

B. Distribution functions

The Keldysh part of Eq. (1) provides the distribution of quasiparticles over energy. $\hat{G}^K$ can be parametrized as $\hat{G}^K=\hat{G}^R\hat{f}-\hat{f}\hat{G}^A$, where the distribution function $\hat{f}$ can be split into parts that are, respectively, odd and even in energy, $\hat{f}=f_L\hat{1}+f_T\hat{\tau}_3$. The kinetic equations for the longitudinal (f_L) and transverse (f_T) distribution

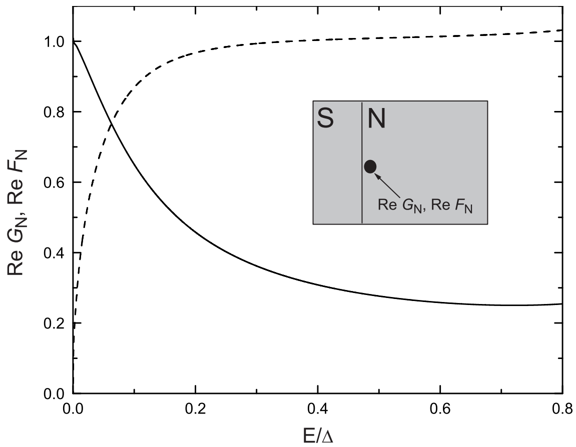

FIG. 2: Calculated density of states (ReG_N, dashed line) and pair amplitude (ReF_N, solid line) as induced by a superconductor into a normal metal, with γ_B=5, γ=0.1, and $d_N \gg \xi_N$. The functions are plotted for the N-side of the interface, as indicated.

functions have the form[24]

$$\nabla\left(D_T \nabla f_T\right) + \operatorname{Im} I_S \nabla f_L = 2 f_T \Delta, \quad (6)$$
$$\nabla\left(D_L \nabla f_L\right) + \operatorname{Im} I_S \nabla f_T = 0, \quad (7)$$

where $D_T = (\operatorname{Re} G)^2 + (\operatorname{Re} F)^2$, $D_L = (\operatorname{Re} G)^2 - (\operatorname{Im} F)^2$ and Δ is the gap inside the superconductor. f_T and f_L determine the quasiparticle and energy flow as can be derived[24] from Eq. (2), giving respectively

$$I_{qp} = \frac{1}{2eR_N} \int dE D_T(E) \nabla f_T(E), \quad (8)$$
$$I_L = \frac{1}{2eR_N} \int dE D_L(E) \nabla f_L(E). \quad (9)$$

The spectral supercurrent is given by $\operatorname{Im} I_S = \frac{1}{8}\operatorname{Tr}[\hat{\tau}_3(\hat{G}^R \nabla \hat{G}^R - \hat{G}^A \nabla \hat{G}^A)] = \operatorname{Im} F^R \operatorname{Re} F^R \nabla \chi$, where χ is the superconducting phase. We consider the regime when superconductivity in the S-wire is not influenced by the normal contacts $N_{1,2}$, which is realized when $\gamma \ll 1$ (large normal-state resistivity of N metal compared to that of S).[23] Then, the product $\operatorname{Im} F_S^R \operatorname{Re} F_S^R$ in S is nonzero only at a narrow energy range near Δ. Since we are interested only in the energy range $E < \Delta$, this allows us to neglect the terms with $\operatorname{Im} I_S \propto \operatorname{Im} F_S^R \operatorname{Re} F_S^R$ in the kinetic equations (6) and (7) at these energies, and the equations for f_T and f_L decouple. The equation for f_T in the wire, consequently, becomes

$$D_T \frac{\partial^2 f_T}{\partial x^2} = 2 f_T \Delta, \quad (10)$$

with the boundary conditions at the NS interfaces given by[25]

$$D_T \gamma_B \frac{\partial}{\partial x} f_T = \pm \operatorname{Re} F \operatorname{Re} F_N \left(f_T - f_{TNi}\right) \quad (11)$$

at $x = \pm d/2$, where $f_{TN}(\pm d/2)$ are the transverse distribution functions in the normal reservours.

Note, that if the assumption $\gamma \ll 1$ is violated, the density of states $\operatorname{Re} G$ in the wire becomes finite at subgap energies. Then, the term $\operatorname{Re} F \operatorname{Re} F_N$ in the above boundary condition will be substituted by $\operatorname{Re} F \operatorname{Re} F_N + \operatorname{Re} G \operatorname{Re} G_N$. However, this will not lead to any qualitative changes in our results and the physical mechanism for the crossed Andreev transport remains the same in this case.

Because of the small barrier transparency, the potential mainly drops at the interfaces and we can assume the electric potential of the S-reservoir and the wire to be zero, and the normal electrodes $N_{1,2}$ to be in equilibrium with the potentials $V_{1,2}$ respectively. Then, the distribution functions in $N_{1,2}$ are

$$f_{Ti,Li} = \frac{1}{2}\tanh\left(\frac{E + eV_i}{2k_B T}\right) \mp \frac{1}{2}\tanh\left(\frac{E - eV_i}{2k_B T}\right).$$

The kinetic equation has the solution $f_T = Ae^{x/\xi} + Be^{-x/\xi}$ where the coherence length ξ is given by $\xi = \sqrt{D_T/2\Delta}$, which describes the conversion of quasiparticle current into supercurrent. This supercurrent is extracted by the S-reservoir at $x = 0$.

III. RESULTS AND DISCUSSION

Solving Eq. (10) with the boundary conditions, Eq. (11), the solution of f_T in the case of two symmetric NS interfaces is given by

$$A = N \frac{f_{T1} e^{-d/2\xi}\left(\gamma_B - N\right) + f_{T2} e^{d/2\xi}\left(\gamma_B + N\right)}{e^{-d/\xi}\left(\gamma_B - N\right)^2 - e^{d/\xi}\left(\gamma_B + N\right)^2},$$
$$B = N \frac{f_{T1} e^{d/2\xi}\left(\gamma_B + N\right) + f_{T2} e^{-d/2\xi}\left(\gamma_B - N\right)}{e^{-d/\xi}\left(\gamma_B - N\right)^2 - e^{d/\xi}\left(\gamma_B + N\right)^2} \quad (12)$$

where $N = \operatorname{Re} F \operatorname{Re} F_N$.

The bias condition provides the final equation from which the distribution functions and currents are derived. The first interface is biased with a voltage V_{bias}, from which f_{T1} is then known. Two bias conditions for the second interface are considered. (a) $I_{qp} = 0$: For a zero total current through the second interface, i.e. an open connection, or an ideal Voltmeter, the nonlocally induced voltage has to be found self-consistently from Eq. (12) and the additional boundary condition,

$$I_{qp} = \int_{-\infty}^{\infty} D_T \frac{\partial}{\partial x} f_T dE = 0.$$

(b) $V_S = V_{N_2}$: Under the alternative bias condition of zero potential difference across the second interface, $f_{T2} = 0$ and Eq. (12) directly provides the solution for f_T in the superconductor.

Note, that it is essential to our approach that $\operatorname{Re} F_N$ is nonzero at subgap energies. When these proximity

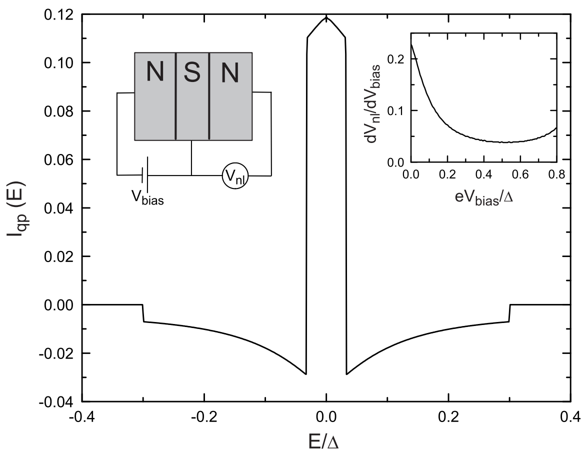

FIG. 3: Spectral quasiparticle current at zero temperature accross the second interface under a voltage bias of the first interface, $V_{\rm bias} = 0.3\Delta$, and a zero total current across the second interface as sketched. The superconducting interlayer thickness $d = 0.3\xi$, and $\gamma_B = 5$. Inset: The response of the induced nonlocal voltage across the second interface (which is the measured quantity of Ref.9) as function of the applied voltage bias across the first interface.

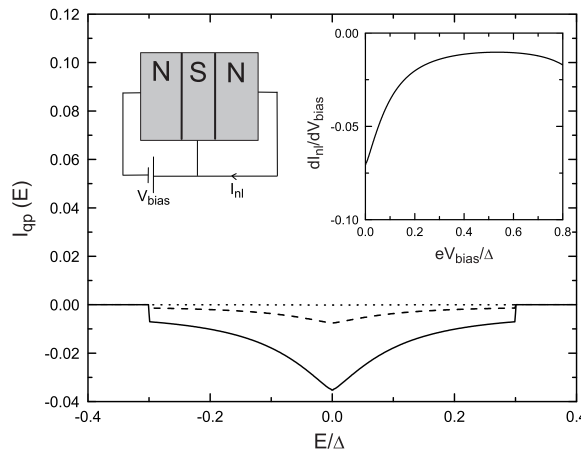

FIG. 4: Nonlocal spectral quasiparticle current at zero temperature across the second interface under a voltage bias of the first interface, $V_{\rm bias} = 0.3\Delta$, and a zero voltage bias of the second interface as sketched. $\gamma_B = 5$ and $d = 0.3\xi$ (solid line), $d = \xi$ (dashed line), $d = 3\xi$ (dotted line). Inset: The response of the total nonlocal current across the second interface as function of the applied voltage bias across the first interface, for $\gamma_B = 5$ and $d = 0.3\xi$.

induced correlations are neglected, the quasiparticle current I_{qp} and the nonlocal effects vanish, coinciding with the results from previous tunnel Hamiltonian[13,17] and circuit theory[19] models.

The resulting spectral quasiparticle currents across the second interface are shown in Figs. 3 and 4, for a thin superconductor of $d = 0.3\xi$ embedded symmetrically between two tunnel barriers with $\gamma_B = 5$. In the case of bias condition (a), a nonlocal voltage, V_{nl}, is induced in the unbiased normal metal electrode, see Fig. 3, as was experimentally observed.[9] V_{nl} is the source of a local Andreev reflection process at the second interface, as characterized by the low-energy peak in $I_{qp}(E)$. In Fig. 1, also $I_{qp}(E)$ across the first interface is shown, to illustrate the different quasiparticle tunneling and reflection processes. A negative current corresponds to a flow of electrons in the positive direction. $I_{qp}(E)$ at the second interface, outside the spectral region of Andreev reflection ($|E| > V_{nl}$) is physically caused by the two nonlocal processes of elastic cotunneling (EC) and crossed Andreev reflection (CAR). The reverse backflow process of EC and CAR from the right to the left electrode results in a suppression of $I_{qp}(E)$ at the first interface for $|E| < V_{nl}$.

In bias situation (b), only the nonlocal currents are contained in $I_{qp}(E)$, see Fig. 4. The magnitude of the nonlocal currents scales exponentially with d/ξ [see solution Eq. (12) and Fig. 4], as expected from the fact that a Cooper pair has size ξ and that nonlocal effects exist by virtue of the coupling to the superconducting condensate. Thus, the Thouless scale is not relevant to our model. The nonlocal currents and voltage scale with γ_B^{-2}, which provides a much larger effect than was found from the tunnel Hamiltonian approach (fourth power in transparency).[13,17]

In the linear response regime (low temperature and low voltage), it was shown recently by Morten *et al.*[19] that

$$\frac{\partial I_{L,qp}}{\partial V_{\rm bias}} = G_{\rm EC}\left(eV_{\rm bias}\right) \pm G_{\rm CAR}\left(eV_{\rm bias}\right), \tag{13}$$

where $G_{\rm EC}$ and $G_{\rm CAR}$ are defined as the nonlocal conductances for EC and CAR respectively. From the fact, that $D_L = 0$ in the superconductor at subgap voltage, and Eq. (9), it follows that $I_L = 0$, and $G_{\rm EC} = -G_{\rm CAR}$. This means that the nonlocal quasiparticle current, $I_{qp}(E) = -\left(G_{\rm EC} - G_{\rm CAR}\right) f_{T1}$, as shown in Fig. 4, is carried by the EC and CAR processes equally, *i.e.* $I_{\rm EC} = I_{\rm CAR} = I_{qp}(E)/2$. The CAR hole in the second electrode can, therefore, be thought of as moving in the negative direction, while the EC electron moves in the positive direction (see Fig. 1).

A treatment of the nonlocal processes in terms of electrons and holes can be derived from the respective electron and hole distribution functions, f_e and f_h, that are given by[24] $f_{h,e} = \left(1 \pm f_L - f_T\right)/2$, when also f_L is treated self-consistently.

The sign and magnitudes of the modeled nonlocal effects are of use for the interpretation of recent experiments,[9,11,12] although many aspects of the experiment related to the geometry are not covered by our model. The obtained equal contribution of EC and CAR to nonlocal currents in NSN structures indicates that additional quasiparticle manipulations are necessary before the device can be considered as a useful source of entangled particles. Creating a non-equilibrium distribution in the electrodes (for example by energy-filtering

in a Fabry-Perot structure) in this respect would be benificial.

IV. CONCLUSION

In summary, within the assumptions of quasiclassical Keldysh-Usadel theory, we find that nonlocal EC and CAR effects exist in a diffusive quasi-1D wire by virtue of the proximity effect. We have found that CAR and EC have the *same* sign in their contribution to the nonlocal spectral quasiparticle current and nonlocal voltage. CAR and EC scale with the square of the barrier transparency, providing large nonlocal effects, of the order of experimentally observed magnitude.

Acknowledgments

Discussions with V. Chandrasekhar, S. Kawabata, T.M. Klapwijk, M.Yu. Kupriyanov, A. Morpurgo, F. Pistolesi, and A.D. Zaikin are gratefully acknowledged. The work was partially supported by the Netherlands Organisation for Scientific Research (NWO) and the NanoNed program under grant TCS7029.

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